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The uncertainty of nitrous oxide emissions from grazed grasslands: A New Zealand case study

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HIGHLIGHTS

- Uncertainty estimated for New Zealand's pastoral agricultural N_2O emissions inventory.
- Emission factor uncertainty estimated by meta-analysis results from 185 field trials.
- Results from new analytic method compared well with Monte Carlo numerical simulation.
- For independent variables and 95% confidence, inventory uncertainty averaged ±58%.

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ABSTRACT

Agricultural soils emit nitrous oxide (N_2O), a greenhouse gas and the primary source of nitrogen oxides which deplete stratospheric ozone. Agriculture has been estimated to be the largest anthropogenic N_2O source. In New Zealand (NZ), pastoral agriculture uses half the land area. To estimate the annual N_2O emissions from NZ's agricultural soils, the nitrogen (N) inputs have been determined and multiplied by an emission factor (EF), the mass fraction of N inputs emitted as N_2O–N. To estimate the associated uncertainty, we developed an analytical method. For comparison, another estimate was determined by Monte Carlo numerical simulation. For both methods, expert judgement was used to estimate the N input uncertainty. The EF uncertainty was estimated by meta-analysis of the results from 185 NZ field trials. For the analytical method, assuming a normal distribution and independence of the terms used to calculate the emissions (correlation = 0), the estimated 95% confidence limit was ±57%. When there was a normal distribution and an estimated correlation of 0.4 between N input and EF, the latter inferred from experimental data involving six NZ soils, the analytical method estimated a 95% confidence limit of ±61%. The EF data from 185 NZ field trials had a logarithmic normal distribution. For the Monte Carlo method, assuming a logarithmic normal distribution for EF, a normal distribution for the other terms and independence of all terms, the estimated 95% confidence limits were −32% and +88% or ±60% on average. When there were the same distribution assumptions and a correlation of 0.4 between N input and EF, the Monte Carlo method estimated 95% confidence limits were −34% and +94% or ±64% on average. For the analytical and Monte Carlo methods, EF uncertainty accounted for 95% and 83% of the emissions uncertainty when the correlation between N input and EF was 0 and 0.4, respectively. As the first uncertainty analysis of an agricultural soils N_2O emissions inventory using "country-specific" field trials to estimate EF uncertainty, this can be a potentially informative case study for the international scientific community.

1. Introduction

In soils, nitrogen (N) containing compounds can be transformed to produce nitrous oxide (N_2O), the third most important greenhouse gas (Davidson and Kanter, 2014). In addition, N_2O is the primary source of nitrogen oxides which deplete stratospheric

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ozone (Ravishankara et al., 2009). Agriculture has been considered the largest source of anthropogenic N_2O emissions and estimated to account for about 70% of the total (Davidson and Kanter, 2014). Pastoral agriculture is practised across 30% of the world's land area, one-third of cropping land area produces animal feed and the world's estimated animal feed N_2O emissions range from 4.4 to 6.8 Tg/y (Herrero et al., 2016). Consequently, the N_2O emissions from pastoral agriculture are substantial, but uncertain.

The N_2O emissions from soils can be attributed to the effects of N inputs (de Klein et al., 2006, 2014a). For pastoral agriculture, the N inputs include N in fertiliser and the urine and dung excreted by grazing animals. The mass fraction of N input emitted from soils as N_2O-N has been denoted an emission factor (EF). Consequently, the N_2O emissions can be estimated by a product of N input and EF. This method includes an inventory of N inputs and EFs following guidelines developed by the Intergovernmental Panel on Climate Change (IPCC, http://www.ipcc-nggip.iges.or.jp/public/2006gl/pdf/4_Volume4/V4_11_Ch11_N2O&CO2.pdf) for signatory nations to the United Nations Framework Convention on Climate Change (UNFCCC).

For the UNFCCC, inventories estimate N_2O emissions across national and annual scales. While N_2O emissions can only be measured at smaller scales, the generalisation of such observations can be the basis for an inventory. Information about the spatial and temporal variability or uncertainty of emission measurements can be informative. For example, 'hot spots' have had a large N input such as urine excreted by grazing animals (Selbie et al., 2015). For grazed areas, hot spots have been found to be the primary source of N_2O emissions (e.g., Kelliher et al., 2002; Giltrap et al., 2014). The temporal variation of N_2O emissions can be dominated by 'hot moments' (Groffman et al., 2009). For example, over 242 days, half the total emissions from grazed pasture comprised three 'events' over 16 days (Scanlon and Kiely, 2003). Following N input, the variability of N_2O emissions has been related to soil wetness which depends on rainfall and irrigation (van der Weerden et al., 2012, 2014).

Ideally, an inventory compiler has accurate and representative information to estimate the emissions. Inventory uncertainty can be estimated by statistically analysing the input information and output calculations (Winiwarter and Muik, 2010). Monte Carlo numerical simulation has been a method used to estimate the uncertainty of agricultural soil N_2O emissions inventories (Winiwarter and Rypdal, 2001; de Vries et al., 2003; Ramirez et al., 2008; Milne et al., 2014). For this method, a set of input data has been determined by sampling from estimated frequency distributions and the inventory computations repeatedly undertaken to calculate an output uncertainty statistic such as 95% confidence limits. Alternatively, provided the inventory can be represented by a suitable mathematical function and input variable statistics estimated, the output uncertainty statistics can be estimated by an analytical method (e.g., Kelliher et al., 2007; Kelliher and Clark, 2010).

Analysing inventory uncertainty can provide information about which factors contribute most to the variability or uncertainty of the output calculations. While as implied, for accuracy and representativeness, the inputs should be determined empirically by a large number of measurements, we are aware of no studies which have taken this approach for a national, annual inventory of agricultural soils N_2O emissions. For those analysing the uncertainty of such inventories, a noticeable stumbling block has apparently been the lack of sufficient "country-specific" EF data. Moreover, to our knowledge, the correlation of N input and EF variables has not been determined empirically due to a lack of sufficient "country-specific" data. In New Zealand (NZ), half the land area is used for pastoral agriculture. Thus, pastoral agriculture is not only vitally important to NZ's economy, it is also a major driver of NZ's greenhouse gas emissions including the agricultural soils N_2O emissions inventory. For these reasons, in NZ, information about pastoral agriculture is important and available. Consequently, predicated on sufficient "country-specific" data, analysing the uncertainty of NZ's agricultural soils N_2O emissions inventory can be a potentially informative case study for the international scientific community.

For this paper, an analytical method will be developed to estimate the uncertainty of an inventory representing NZ's agricultural soil N_2O emissions. Estimates will also be made using the Monte Carlo method and results from the two methods will be compared. While estimating N input uncertainty will have to rely on expert judgement, EF uncertainty will be estimated by meta-analysis of the results from 185 NZ field trials (Kelliher et al., 2014a). As will be shown, while a correlation between N input and EF affects the inventory's uncertainty, inventory-scale data were not available to estimate the correlation. Instead, the correlation will be inferred by meta-analysis of replicate-level results from experiments involving six NZ soils (de Klein et al., 2014b; Kelliher et al., 2014b; Venterea et al., 2015). The estimated uncertainty of NZ's inventory will be compared with estimates for inventories from other countries.

2. Material and methods

The N_2O emissions from agricultural soils (E_{N_2O}) can be estimated by a product of N input and a representative value of EF. For N input, we need to estimate the annual mass of N returned by the excreta of grazing animals which includes dairy and beef cattle, sheep and deer. Thus, we need to determine the number of animals, a, and mean values of the animal's annual energy requirement (d, MJ animal^{-1} y^{-1}), the pasture (feed) energy content (e, MJ kg^{-1} [dry matter] = MJ kg^{-1} DM), the pasture N content, p_N, and the fraction of N that will be retained in an animal, r_N. We combined these terms as $\left[a\,d\left(\frac{1}{e}\right)p_N(1-r_N)\right]$. In addition, we need to estimate the annual mass of N fertiliser applied to agricultural soils across NZ, most commonly as urea and denoted by term u. An E_{N_2O} equation was then be written as:

$$E_{N_2O} = \left\{\left[a\,d\left(\frac{1}{e}\right)p_N\,(1-r_N)\right]+u\right\}\left(\frac{44}{28}\right)EF \qquad (1)$$

Using the N_2O/N_2 molecular mass ratio (44/28 = 1.57), units on the right hand side of the equation were converted from Gg N y^{-1} and E_{N_2O} expressed as Gg N_2O y^{-1}.

For uncertainty analysis, we used data from NZ's E_{N_2O} inventory for the year 2014 (Ministry for the Environment, 2016). As shown below, the quantity ($a\,d$) needs to be 585 × 10^9 MJ y^{-1}. For this purpose, the mean values of p_N, r_N and e will be 0.035 (kg N kg^{-1} DM), 0.15 and 11 MJ kg^{-1} DM, respectively (Kelliher et al., 2007). By inserting these 4 values into $\left[a\,d\left(\frac{1}{e}\right)p_N(1-r_N)\right]$, we calculated 1582 Gg N y^{-1} the value used for NZ's 2014 inventory. Based on N fertiliser sales, u will be 377 Gg N y^{-1} (Ministry for the Environment, 2016). The NZ inventory also accounts for E_{N_2O} from managed excreta attributed to dairy cattle during milking, crop residues and cultivated organic soils. For the year 2014, these totalled 1.5 Gg N_2O (Ministry for the Environment, 2016). This quantity will be added to the result of the calculation using Equation (1).

To determine a representative *EF* value, we combined the urine,

dung and N fertiliser EFs from NZ's inventory and estimated a weighting factor for each EF. The urine, dung and N fertiliser EFs were 0.01, 0.0025 and 0.0048 kg N_2O N/kg N input, respectively. Based on NZ's 2014 inventory, we determined that urine was 67% of the excreta N and dung was 33%. Also based on NZ's 2014 inventory, excreta comprised 81% of the total N applied to soils, while N fertiliser comprised 19%. Thus, the weighting factor was 0.543 (0.67 × 0.81) for urine, 0.267 for dung and 0.19 for N fertiliser and the weighted mean *EF* was 0.007 (Table 1).

The inventory also accounts for the emissions from managed excreta, crop residues and cultivated organic soils. In addition, the inventory accounts for indirect emissions of N_2O that come from a proportion of N that volatilises as ammonia (10%), and will be re-deposited onto soils, and for another proportion of N which leaches through soils (7%). For uncertainty analysis, as shown below, we will account for these emissions by firstly adding a factor to the direct emissions. Then, we will multiply our estimate of E_{N_2O} by another factor determined by the value which yields the inventory's sum of the direct and indirect emissions, the total emissions. There was no uncertainty information about the emissions from managed excreta, crop residues and cultivated organic soils or the indirect emissions, so no uncertainty will be assigned to these factors.

The terms in Equation (1) were mean values based on sets of imperfect measurements or judgements. The uncertainty of each value was quantified by the fractional standard error (*FSE*, Tables 2 and 3). As an example, for term *d*, the notation was $FSE[d] = SE[d]/\mu_d$ where μ_d denoted the mean value of *d*. To determine *FSE*[*EF*], the weighting factors were used again. The corresponding *FSE* values were determined by a meta-analysis of results from 185 field trial across NZ (Kelliher et al., 2014a,b). For example, on average, the FSE for urine was 0.276 based on 91 field trials at lowland and hill country, low slope sites (Table 2). The *FSE* values were the statistical basis for an analytical method to estimate the uncertainty of NZ's E_{N_2O} inventory. This method has been developed in an Appendix.

3. Results

3.1. NZ's E_{N_2O} inventory

Using Equation (1), we estimated NZ's 2014 E_{N_2O} inventory. Thus, we inserted the values for $\left[a\ d\ \left(\frac{1}{e}\right)p_N(1-r_N)\right]$ (=1582 Gg N y^{-1}) and *u* (=377 Gg N y^{-1}), multiplied by the weighted mean *EF* (=0.007) and by 1.57 to obtain 21.5 Gg N_2O y^{-1}. Next, to account for the emissions from managed excreta, crop residues and cultivated organic soils, we added 1.5 Gg N_2O y^{-1} which gave 23.0 Gg N_2O y^{-1} as the estimated direct N_2O emissions. To complete the calculation of total N_2O emissions, we need to include the indirect N_2O emissions. By the IPCC method used by NZ for UNFCCC reporting, the total N_2O emissions from agricultural soils for 2014 was 28.6 Gg N_2O y^{-1} (Ministry for the Environment, 2016). Thus, a ratio of the total and estimated direct N_2O emissions was 1.23 (= 28.6/23.0). Consequently, we multiplied the estimated direct N_2O emissions by 1.23 to determine the total N_2O emissions as 28.6 Gg N_2O y^{-1}.

Table 1
Component weighting factors and corresponding mean values of *EF* from New Zealand's E_{N_2O} inventory which were used to calculate a weighted mean *EF* for an equation in the text and equations in the Appendix used to estimate the uncertainty of NZ's E_{N_2O} inventory.

Nitrogen source	Weighting factor	*EF*
Urine	0.543	0.01
Dung	0.267	0.0025
Urea fertiliser	0.190	0.0048
Weighted mean *EF*		0.0070

Table 2
Fractional standard errors (*FSE*) from Kelliher et al. (2014a) and the component weighting factors which were used to calculate the weighted mean *FSE* of term *EF* for an equation in the text and equations in the Appendix used to estimate the uncertainty of NZ's E_{N_2O} inventory.

Nitrogen source	Weighting factor	*FSE*
Urine	0.543	0.276
Dung	0.267	0.282
Urea fertiliser	0.190	0.271
Weighted mean *FSE*		0.277

Table 3
Fractional standard errors (*FSE*) for the terms in Equation (1) and Equation (A1) in the Appendix. The FSE for term r_N was determined by the FSE for term *e* on the basis of CSIRO (2007, see Equation (1.12A)). The *FSE* values are the statistical basis for estimating the uncertainty of New Zealand's E_{N_2O} inventory as explained in the Appendix.

Term in Equation (1)	*FSE*	Method used to estimate *FSE*
a	0.02	Statistics NZ (Kelliher et al., 2007)
d	0.05	Expert judgement (Kelliher et al., 2007)
e	0.05	Expert judgement (Kelliher et al., 2007)
p_N	0.01	Expert judgement (Ledgard et al., 2002)
r_N	0.05	Expert judgement
u	0.03	Expert judgement (Hilton Furness, pers. comm.)
EF	0.277	Statistical analysis of the data in Tables 1 and 2

3.2. E_{N_2O} uncertainty by an analytical method assuming N input and EF were independent

To begin estimating the uncertainty of NZ's E_{N_2O} inventory, we assumed independence of the terms in Equation (1) and equations in the Appendix. On this basis, in the Appendix, we developed Equation (A3) to calculate an *FSE* for the annual mass of N excreta returned by grazing animals to agricultural soils, denoted by term *x* and its associated *FSE*[*x*]. By estimating r_N as 0.15 and using the 5 *FSE* values from Table 3, we calculated *FSE*[*x*] was 0.075. We then used Equation (A4), and by inserting *FSE*[*x*] of 0.075, *x* of 1582 Gg N y^{-1}, *FSE*[*u*] of 0.03, *u* of 377 Gg N y^{-1} and *FSE*[*EF*] of 0.277, we calculated $FSE[E_{N_2O}]$ was 0.284. Consequently, 95% of $FSE[E_{N_2O}]^2$ was attributed to $FSE[EF]^2$ (=[0.277^2/0.284^2]*100). As stated, including the indirect N_2O emissions, the total E_{N_2O} was 28.6 Gg N_2O y^{-1}, so $SE[E_{N_2O}]$ was 8.1 Gg N_2O y^{-1} (= 28.6*0.284). For a 95% confidence limit, we required twice $SE[E_{N_2O}]$ or 16.2 Gg N_2O y^{-1}. Thus, by these calculations and assuming a normal distribution, we were 95% certain that the true value of NZ's E_{N_2O} was 28.6 ± 16.2 Gg N_2O y^{-1} (±57%) or between 12.4 and 44.8 Gg N_2O y^{-1}, a (95% confidence) range of 32.4 Gg N_2O y^{-1}.

3.3. E_{N_2O} uncertainty by an analytical method assuming N input and EF were correlated

If the terms representing NZ's inventory were correlated, not independent, the $FSE[E_{N_2O}]$ estimate would be different, depending on the degree of correlation. To examine the effects of this assumption, we constructed and analysed another, simpler representation of the inventory. As described in the Appendix, this involved subsuming the terms related to *x* and *u* into a single term

denoted *N input*. Then, to determine *FSE*[*N input.EF*] using the Appendix's culminating Equation (A15), we required estimates of *FSE*[*N input*], *FSE*[*EF*] and $\rho_{N\ input,\ EF}$. In Table 2, we showed *FSE*[*EF*] = 0.277 and using the data in Table 3 with Equations (A3) and (A4), *FSE*[*N input*] = 0.061. To illustrate by a sensitivity analysis, we calculated *FSE*[*N input.EF*] for maximum and minimum values of $\rho_{N\ input,\ EF}$. These calculations were then compared to the $FSE[E_{N_2O}]$ or *FSE*[*N input.EF*] estimate of 0.284 obtained earlier, assuming $\rho_{N\ input,\ EF}$ was 0 (from independence of *N input* and *EF*). Thus, if *FSE*[*N input*] and *FSE*[*EF*] were 0.061 and 0.277, respectively, and $\rho_{N\ input,\ EF}$ was +1 and -1, *FSE*[*N input.EF*] was 0.332 (17% larger) and 0.222 (23% smaller), respectively.

To estimate $\rho_{N\ input,\ EF}$ for NZ's inventory, we analysed replicate-level *N input* and *EF* data from field and laboratory experiments involving six NZ soils (de Klein et al., 2014b; Kelliher et al., 2014b; Venterea et al., 2015). Briefly, for each soil, there were four to six levels of N input from 100 to 1500 kg N/ha, three to five replicates per level of N input and $\rho_{N\ input,\ EF}$ was estimated by linear regression. The mean result was 0.40 ± 0.37 for 95% confidence (Table 4). Inserting $\rho_{N\ input,\ EF}$ of 0.40, *FSE*[*N input*] of 0.061 and *FSE*[*EF*] of 0.277 into Equation (A15), we estimated *FSE*[*N input.EF*] was 0.304 (7% larger than 0.284 when $\rho_{N\ input,\ EF}$ was 0). Therefore, 83% of $FSE[E_{N_2O}]^2$ was attributed to $FSE[EF]^2$ (=$[0.277^2/0.304^2]$*100). Moreover, $FSE[E_{N_2O}]$ became 8.7 Gg N_2O y^{-1} (= 28.6*0.304) and assuming a normal distribution the 95% confidence limit was 28.6 ± 17.4 Gg N_2O y^{-1} (±61%).

3.4. E_{N_2O} uncertainty by the Monte Carlo method

The *FSE* values in Tables 2 and 3 were also used as the statistical basis for the Monte Carlo method to estimate the uncertainty of NZ's E_{N_2O} inventory. A normal distribution was chosen to represent each variable's distribution with one exception because there was sufficient evidence to justify a different choice. For EF, the results of 185 NZ field trials (Kelliher et al., 2014a) indicated this variable should be represented by a log normal distribution (Fig. 1). The simulation used the same values for $\left[a\ d\left(\frac{1}{e}\right)p_N(1-r_N)\right]$, the weighted mean *EF* and corresponding FSE (Tables 1 and 2) and the calculations described earlier to determine E_{N_2O}. Assuming the independence of variables, the simulation indicated 95% certainty that a true value of E_{N_2O} was between 16.8 and 46.9 Gg y^{-1}, a (95% confidence) range of 30.1 Gg y^{-1}. The corresponding mean and median E_{N_2O} values and $FSE[E_{N_2O}]$ were 24.9 and 27.8 Gg y^{-1} and 0.284, respectively. Thus, the simulation's mean was 12% less than NZ's E_{N_2O} inventory value of 28.6 Gg y^{-1} and the median was 3% less. Using the simulation's mean as done for the analytical method, the estimated uncertainty was -32% (= 100 − [100 {16.8/24.9}]), +88% (= [100 {46.9/24.9}] - 100) and ±60% on average with asymmetry due to the log normal distribution for EF. We also estimated 95% of $FSE[E_{N_2O}]^2$ was attributed to $FSE[EF]^2$ (= $[0.277^2/0.284^2]$*100), the same percentage estimated by the analytical method when the variables had been assumed to be independent. Alternatively, if the variables were positively correlated with a correlation coefficient of 0.40, the estimated $FSE[E_{N_2O}]$ became 0.304 and the inventory's estimated uncertainty became -34%, +94% and ±64% on average. Moreover, as for the analytical method, 83% of $FSE[E_{N_2O}]^2$ was attributed to $FSE[EF]^2$ (= $[0.277^2/0.304^2]$*100).

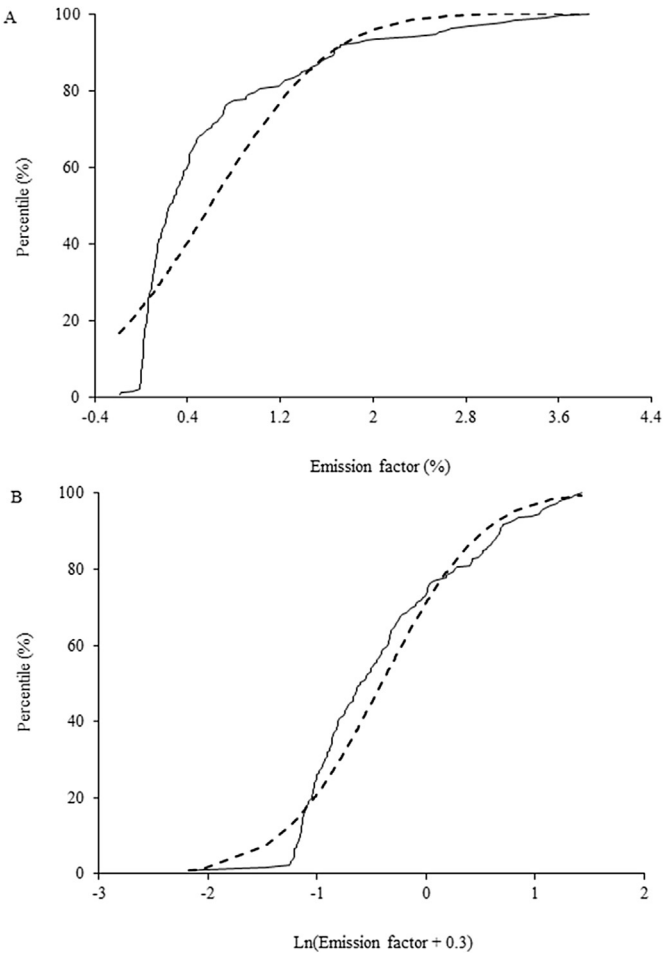

Fig. 1. Cumulative frequency distribution of (A) (raw) mean emission factor data from 185 NZ field trials which included cattle and sheep urine and dung and urea fertiliser (solid line, Kelliher et al., 2014a) and (B) the same data after adding a constant of 0.3 and (base e) logarithmic transformation. The dashed lines are normal distributions calculated using the data's overall mean and standard deviation.

Table 4
Correlation of N input and N_2O EF for six NZ soils determined by meta-analyses of the replicate data from laboratory incubation studies (Kelliher et al., 2014b; Venterea et al., 2015) and field trials (de Klein et al., 2014a,b).

Soil	Correlation	Source
Temuka	0.00	Kelliher et al. (2014b)
Horotiu	+0.50	de Klein et al. (2014b)
Te Kowhai	0.00	de Klein et al. (2014b)
Wingatui	+0.43	de Klein et al. (2014b)
Wakanui	+0.94	Venterea et al. (2015)
Waikari	+0.52	Venterea et al. (2015)
Mean	+0.40	

4. Discussion

EF uncertainty was the principal determinant of E_{N_2O} uncertainty for NZ's inventory. The EF uncertainty was estimated by meta-analysis of the results from 185 NZ field trials. An estimated 95% and 83% of the uncertainty of NZ's E_{N_2O} inventory was attributed to EF uncertainty when the correlation between N input and EF was 0 and 0.4, respectively. The meta-analysis also provided sufficient evidence to justify representing the EF uncertainty using a logarithmic normal distribution. This was done using the Monte Carlo method and the average percentage estimate of E_{N_2O} uncertainty was similar to that by the analytical method which had

assumed a normal distribution for the EF uncertainty. However, the Monte Carlo method yielded asymmetrical estimates of E_{N_2O} uncertainty equal to −32% and +88% and −34% and +94% when the correlation between N input and EF was 0 and 0.4, respectively. This has been the first E_{N_2O} inventory uncertainty analysis to estimate EF uncertainty by meta-analysis of results from "country-specific" field trials. However, we acknowledge the inference from field trials to national annual inventory was made across considerable time and space scales. This also applied to the estimated correlation between N input and EF. For those field and laboratory experiments, the N input to a plot was increased by increasing N concentration of the applied aqueous solution. Across larger scales for NZ's inventory, we recognize that N input can also be increased by increasing the number of grazed areas (paddocks) and paddocks receiving N fertiliser.

To our knowledge, previous accounting for the effect of $\rho_{N\ input,\ EF}$ on inventory E_{N_2O} uncertainty has been by sensitivity analysis using the Monte Carlo method (eg, Ramirez et al., 2008). Alternatively, as stated, we analysed replicate-level *N input* and *EF* data from field and laboratory experiments involving six NZ soils. Under field conditions, $\rho_{N\ input,\ EF}$ was similar for two soils studied by de Klein et al. (2014b) with values about the same as the overall mean, but for unknown reasons, $\rho_{N\ input,\ EF}$ was zero for a third soil. Under field conditions, *N input* to soils can be affected by plant N uptake (Ma et al., 2010). Instead, though artificial, conditions can be controlled in a laboratory and soil samples incubated without plants. Under the laboratory conditions studied by Kelliher et al. (2014b), $\rho_{N\ input,\ EF}$ was zero for a Temuka soil sampled near Lincoln. Under the laboratory conditions studied by Venterea et al. (2015), $\rho_{N\ input,\ EF}$ was 0.94 for a Wakanui soil sampled near Lincoln (about 3 km from where the Temuka soil had been sampled, T.J. Clough, personal communication) and 0.52 for a Waikari soil sampled about 90 km away. The Waikanui and Waikari soils were chosen for their similar texture, pH, carbon content and carbon to N ratio. For a given *N input*, the Wakanui soil had a mean EF up to six time larger than that of the Waikari soil. In response to *N input*, the Waikari soil was more resistant to nitrite accumulation and the abundance of microbial genes associated with nitrite oxidation increased substantially (Venterea et al., 2015). While the $\rho_{N\ input,\ EF}$ results for six NZ soils were variable, Venterea et al. (2015) have developed a useful hypothesis for better understanding the results of future studies.

The considerable uncertainty of NZ's E_{N_2O} inventory warrants comparison. For the Monte Carlo method, assuming a logarithmic normal distribution for EF, a normal distribution for the other terms and independence of all terms, the estimated 95% confidence limits were −32% and +88% or ±60% on average. For Finland with an E_{N_2O} inventory calculated using the IPCC method and uncertainty assessed by the Monte Carlo method, the estimated 95% confidence limits were −52% and +70% or ±61% on average (Monni et al., 2007). In contrast, using a similar approach, the estimated 95% confidence limit for the Netherlands E_{N_2O} inventory averaged ±42% (Ramirez et al., 2008). Moreover, also using a similar approach, the estimated 95% confidence limits for the United Kingdom's (UK's) E_{N_2O} inventory were −56% and +140% or ± 98% on average (Milne et al., 2014).

As stated, EF uncertainty was the principal determinant of E_{N_2O} uncertainty for NZ's inventory. Because EF uncertainty has been attributed to soil wetness, Monni et al. (2007) argued that EF uncertainty might be reduced by estimating EF using climate data. For example, spatially- and temporally-disaggregated EFs (500 by 500 m^2 and daily, respectively) were estimated for dairy cattle urine across NZ using a predictive relationship with soil water content and a water balance model implemented using a geographic information system (van der Weerden et al., 2014). However, the uncertainty of this calculation methodology has not been assessed. For the Netherlands, an E_{N_2O} inventory was calculated using a hydrology and N flow model driven by rainfall and N input to soils (de Vries et al., 2003). The model's parameters were estimated from relationships with soil use, soil type and groundwater level class derived from 500 by 500 m^2 grid cell data on digital maps. For this inventory, the estimated 95% confidence limit was ±48% (±twice the standard deviation as a percentage of the mean which was reported in their Table 17, de Vries et al., 2003). Kros et al. (2012) also calculated an E_{N_2O} inventory for the Netherlands using a spatial (1 km^2 grid cells) model and the estimated 95% confidence limit was ±38%. Kros et al. (2012) reckoned de Vries et al. (2003) had over-estimated the inventory's uncertainty by "ignoring spatial correlation", a criticism which can also be applied to the IPCC methodology. Kros et al. (2012) accounted for spatial correlation by using their spatial model and Equation (A13) developed by Lesschen et al. (2007). While spatial and IPCC methods used to estimate E_{N_2O} inventories for the Netherlands have corresponded with some differences in uncertainty assessment, the estimated percentage uncertainties reported by Kros et al. (2012) and Ramirez et al. (2008) were notably similar.

The effects of uncertainties such as climate change and livestock production in future have not been determined by this study. While estimating the consequences of climate change on the N_2O emissions from agricultural soils remains uncertain, case studies for NZ (de Klein et al., 2014a) and the UK (Abalos et al., 2016) suggest climate change should be expected to correspond with increased emissions in future. Likewise, in future, the global demand for animal products such as meat should be expected to increase (e.g., Thornton, 2010). Increased demand could be met by more animals and/or increased feed which should also correspond with increased emissions in future. However, we must be cautious about whether or not even likely time trends will substantially affect the uncertainty of NZ's E_{N_2O} inventory.

5. Conclusions

The uncertainty of NZ's E_{N_2O} inventory could be estimated by statistically evaluating the input information and output calculations by analytical and Monte Carlo methods. This required inference from the input information across considerable time and space scales. While the inventory had to be simplified for representation, choosing a product of *N input* and *EF* was defensible. The subsequent development of a suitable set of mathematical functions for the analytical method was enabled by Goodman's (1960) exact expression for the variance of a product. Using the analytical method's culminating function, the inventory's uncertainty could be estimated for all levels of correlation between the primary variables. Results from the analytical and Monte Carlo method calculations were similar. EF uncertainty was the principal determinant of E_{N_2O} uncertainty for NZ's inventory. An estimated 95% and 83% of the uncertainty of NZ's E_{N_2O} inventory was attributed to EF uncertainty when the correlation between N input and EF was 0 and 0.4, respectively. For a mean correlation of 0.40, the inventory's uncertainty increased by an estimated 7% compared to no (zero) correlation. For the Monte Carlo method, assuming a logarithmic normal distribution for EF, a normal distribution for the other terms and independence of all terms, the estimated 95% confidence limits were −32% and +88% or ±60% on average.

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Appendix

The N_2O emissions from agricultural soils (E_{N_2O}) can be attributed to the effects of nitrogen (N) input. To proceed, we need to estimate the annual mass of N returned by the excreta of grazing animals which includes dairy and beef cattle, sheep and deer. This requires us to determine the number of animals, a, and mean values of the animal's annual energy requirement (d, MJ animal^{-1} y^{-1}), the pasture (feed) energy content (e, MJ kg^{-1} [dry matter] = MJ kg^{-1} DM), the pasture N content, p_N, and the fraction of N that will be retained in an animal, r_N. We can combine these terms as $\left[a\,d\left(\frac{1}{e}\right)p_N(1-r_N)\right]$. For analysis, we will combine these terms into an excreta factor called x as

$$x = a\,d\left(\frac{1}{e}\right)p_N\,(1-r_N) \tag{A1}$$

In addition, we need to estimate the annual mass of N fertiliser applied to agricultural soils across NZ, most commonly as urea and denoted by term u. An emission factor (EF) is the mass fraction of N *input* emitted as N_2O-N to the atmosphere. Because we can estimate E_{N_2O} by a product of N *input* and EF, we can write a simple equation to represent NZ's E_{N_2O} inventory as:

$$E_{N_2O} = (x+u)\left(\frac{44}{28}\right)EF \tag{A2}$$

where the N_2O/N_2 molecular mass ratio (44/28) converts units from Gg N y^{-1} to Gg N_2O y^{-1}.

The terms in equations (A1) and (A2) will be mean values based on sets of imperfect measurements or judgements. The uncertainty of each value will be quantified by the fractional standard error FSE(Tables 2 and 3). As an example, for term d, the notation will be $FSE[d] = \frac{SE[d]}{\mu_d}$ where μ_d denotes the mean value of dWe can determine $FSE[x]$ by a root-mean-square approach, recognizing the mathematical operation involving term r_N, as

$$FSE[x]^2 = FSE[a]^2 + FSE[d]^2 + FSE[e]^2 + FSE[p_N]^2 + \left\{FSE[r_N]\frac{r_N}{1-r_N}\right\}^2 \tag{A3}$$

where we have written $FSE[x]^2$ to denote $(FSE[x])^2$. To assess the uncertainty of E_{N_2O}, we can write the following approximation

$$FSE\,[E_{N_2O}] \approx \left\{\left(\frac{x^2FSE\,[x]^2 + u^2FSE\,[u]^2}{(x+u)^2}\right) + FSE[EF]^2\right\}^{0.5} \tag{A4}$$

An assumption of independence has been made for the terms in Equations (A2)–(A4) in order to estimate $FSE[E_{N_2O}]$. If the terms had been correlated, the estimate of $FSE[E_{N_2O}]$ would have been different, depending on the degree of correlation. To introduce the effects of this assumption, we will construct and analyse another, simpler representation of the inventory. This will involve subsuming $\left\{\left[a\,d\left(\frac{1}{e}\right)p_N\,(1-r_N)\right]+u\right\}\left(\frac{44}{28}\right)$ into a single term denoted N *input*, so Equation (A2) can be re-written explicitly as the product of N *input* and EF

$$E_{N_2O} = N\ input.EF \tag{A5}$$

Assuming independence of the terms in Equation (A5), we would again follow a root-mean-square approach and write the following approximation

$$FSE[E_{N_2O}] \approx \left(FSE[N\ input]^2 + FSE[EF]^2\right)^{0.5} \tag{A6}$$

This approximation follows from an exact expression for the product of independent terms which is given by

$$FSE[E_{N_2O}] = \left(FSE[N\,input]^2 + FSE[EF]^2 + FSE[N\,input]^2FSE[EF]^2\right)^{0.5} \tag{A7}$$

When $FSE[N\ input]$ and/or $FSE[EF]$ are small, $FSE[N\ input]^2FSE[EF]^2$ will be very small and can be ignored. Based on Equation (A7) and data given in the paper and Tables 2 and 3, it can be shown $FSE[N\ input] = \left(\frac{x^2FSE[x]^2+u^2FSE[u]^2}{(x+u)^2}\right)^{0.5} = 0.061$ and $FSE[EF] = 0.277$, so $FSE[N\ input]^2FSE[EF]^2 = 0.00029$. Ignoring this small quantity, the approximation given by Equation (A6) is shown to be adequate, yielding $FSE[E_{N_2O}] = 0.284$, the same estimate which can be calculated by equation (A7). However, if N *input* and EF had been correlated, another (exact) expression would be needed. The seminal study of Goodman (1960) provides such an expression that will be the basis for our next analysis.

An exact expression for the variance (denoted var) of a product was developed by Goodman (1960) and we write

$$\begin{aligned} var[N\ input.EF] = {} & var[N\ input]var[EF] + \left(\mu_{N\ input}\right)^2 var[EF] \\ & + (\mu_{EF})^2 var[N\ input] + cov\left[N\ input^2,\ EF^2\right] \\ & - (cov[N\ input,\ EF])^2 \\ & - 2\mu_{N\ input}\mu_{EF}cov[N\ input,\ EF] \end{aligned} \tag{A8}$$

where $\mu_{N\ input}$ is the mean of N *input*, μ_{EF} is the mean of EF and the $cov[N\ input, EF]$ is the covariance of N *input* and EF.

Given $cov[N\ input^2, EF^2]$ is $4\mu_{N\ input}\mu_{EF}cov[N\ input, EF]$, based on a Taylor Series, Equation (A8) may be approximated by

$$\begin{aligned} var[N\ input.EF] \approx {} & var[N\ input]var[EF] + \left(\mu_{N\ input}\right)^2 var[EF] \\ & + (\mu_{EF})^2 var[N\ input] - (cov[N\ input, EF])^2 \\ & + 2\mu_{N\ input}\mu_{EF}cov[N\ input, EF] \end{aligned} \tag{A9}$$

The approximation includes two terms involving the covariance of N *input* and EF. One is subtracted and the other depends on the sign of the covariance (when the means are positive). To proceed, we briefly re-consider the situation when N input and EF are independent, such that Equations (A8) and (A9) can be reduced to the first three terms as

$$var[N\ input.EF] = var[N\ input]var[EF] + \left(\mu_{N\ input}\right)^2 var[EF] + (\mu_{EF})^2 var[N\ input] \quad (A10)$$

Equation (A10) can be rearranged using an expression for $FSE[N\ input] = \frac{SE[MN\ input]}{\mu_M}$ and $SE[N\ input] = (var[N\ input])^{0.5}$ and $\mu_{N\ input.\ EF} = \mu_{N\ input}\mu_{EF}$ (because of independence) to give FSE of the product [*N input.EF*] as

$$FSE[N\ input.EF] = \frac{SE[N\ input.EF]}{\mu_{N\ input.EF}} = \frac{SE[N\ input.EF]}{\mu_{N\ input}\mu_{EF}}$$

which can be expanded, as before for Equation (A7), and repeated here in a different order as

$$FSE[N\ input.EF] = \left(FSE[N\ input]^2 FSE[EF]^2 + FSE[EF]^2 + FSE[N\ input]^2\right)^{0.5} \quad (A11)$$

Now, when *N input* and *EF* are not independent, the covariance of *N input* and *EF* can be expressed in terms of the correlation of *N input* and *EF*, $\rho_{N\ input,\ EF}$, which can be written as $cov[N\ input, EF] = \rho_{N\ input,\ EF} SE[N\ input]SE[EF]$. We can use this expression to re-write Equation (A9) as

$$var[N\ input.EF] \approx var[N\ input]var[EF] + \left(\mu_{N\ input}\right)^2 var[EF] + (\mu_{EF})^2 var[N\ input] - \left(\rho_{N\ input,EF} SE[N\ input]SE[EF]\right)^2 + 2\mu_{N\ input}\mu_{EF}\rho_{N\ Input,EF} SE[N\ input]SE[EF] \quad (A12)$$

From Equation (A12), when *N input* and *EF* are positively correlated with positive means, *var*[*N input.EF*] will be greater than that given by Equation (A10) for independent *N input* and *EF* when $\frac{2\mu_{N\ input}\mu_{EF}}{SE[N\ input]SE[EF]} > \rho_{N\ input,\ EF} > 0$. Rearranging the left inequality gives $2 > \rho_{N\ input,\ EF} FSE[N\ input]FSE[EF]$, which will certainly be satisfied when *FSE*[*N input*] and *FSE*[*EF*] are less than 1. When *N input* and *EF* are negatively correlated (with positive means), *var*[*N input.EF*] will be less than that in Equation (A10) because the last 2 terms in Equation (A12) will be negative. Further, when *N input* and *EF* are correlated

$$\mu_{N\ input.EF} = \mu_{N\ input}\mu_{EF} + cov[N\ input, EF] = \mu_{N\ input}\mu_{EF} + \rho_{N\ input,EF} SE[N\ input]SE[EF] \quad (A13)$$

and

$$\frac{\mu_{N\ input.EF}}{\mu_{N\ input}\mu_{EF}} = 1 + \rho_{N\ input,EF}\frac{SE[N\ input]SE[EF]}{\mu_{N\ input}\mu_{EF}} = 1 + \rho_{N\ input,EF} FSE[N\ input]FSE[EF] \quad (A14)$$

We can use Equation (A12) and (A14) to derive a general expression for *FSE*[*N input.EF*]. This will recognize that $FSE[N\ input.EF] = \frac{SE[N\ input.EF]}{\mu_{N\ input.EF}}$. Moreover, from Equation (A14) when *N input* and *EF* are correlated, we know $FSE[N\ input.EF] = \frac{SE[N\ input.EF]}{\mu_{N\ input}\mu_{EF}}\frac{\mu_{N\ input}\mu_{EF}}{\mu_{N\ input.EF}}$. Thus, for all levels of correlation between *N input* and *EF* including negative, positive and nil (independence), we can now write

$$FSE[N input.EF] \approx \frac{\left\{FSE[N input]^2 FSE[EF]^2 + FSE[EF]^2 + FSE[N input]^2 - \left(\rho_{N input,EF} FSE[N input]FSE[EF]\right)^2 + 2\rho_{N input,EF} FSE[N input]FSE[EF]\right\}^{0.5}}{\left(1 + \rho_{N input,EF} FSE[N input]FSE[EF]\right)} \quad (A15)$$

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